MAKING VIABLE THE RECYCLING OF CARBON FIBER/THERMOSET MATRIX COMPOSITES. FIRST ELEMENTS OF STUDY

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ABSTRACT

Originally developed for high-tech applications, carbon/epoxy composites have been increasingly used in leisure and sports industries, for several years. Nevertheless, the carbon reinforcement is an expensive constituent, and it has been recently shown that it is also the most environmentally impacting in a composite part manufacturing. In this way, recycling these materials (even restricted to the reinforcement recovery) could lead to reduce economic and environmental inadequacies, while satisfying legislative requirements for their end-of-life.

This article is the basis for a life cycle analysis that assesses benefits and environmental challenges of this recycling loop. The recovery of the carbon reinforcement (based on the solvolysis of the matrix by water under supercritical conditions) offers an average gain of 70% for all eco-indicators.

INTRODUCTION

Carbon fiber/thermoset matrix composites were originally developed for high-tech applications in aeronautic and aerospace fields. However for several years, these materials have been increasingly used in automotive and leisure and sports industries. In these sectors, one may seek aesthetic criterions or a simple feeling of high technology, more than high technical properties. Thus, constituents' characteristics, and specifically reinforcement's (*e.g.* Young's modulus, tensile strength, etc.), are considered as a secondary matter and may be oversized regarding the function of the product. This is particularly true for non-structural decorative parts (*e.g.* with a carbon look finish), for which the reinforcement is the most expensive constituent, and where glass fibers, very much cheaper, cannot be used.

As a consequence for these scopes, alternative solutions to technical and economic inadequacies have to be found, but kept in line with the environmental impact of the product. Indeed, it has been shown that the carbon reinforcement is environmentally the most impacting constituent in a carbon/epoxy composite's elaboration process (Duflou, De Moor, Verpoest, & Dewulf, 2009) (cf. analysis Figure 1). First, recycling end-of-life composites (even restricted to the reinforcement recovery) could lead to reduce some anthropogenic impacts, by decreasing the use of first-generation raw materials (mainly petroleum) for their production. Besides, it would help design engineers to balance energy efficiency and cost, by opening new opportunities for developing second-generation composites, firstly dedicated to the manufacture of medium or low loaded parts. Lastly, recycled carbon fabric could widen the range of reinforcements on the marketplace, between first-generation carbon and glass fibers.

All this has to be led in line with European directives that already force industries to improve their products' recyclability (*e.g.* in automotive industry (European Parliament & Council of the European Union, 2000, p. 53)). However, making viable this new recycling sector requires overcoming users' reluctances by ensuring the second-generation semi-product's validity from economic and environmental aspects. Therefore, we carried out a life cycle assessment (LCA), in which the resource efficiency and potential environmental challenges of the carbon/epoxy composites' recycling process are analyzed.

MATERIALS AND METHODS

To assess a LCA, every stage of the life cycle of the composite part has to be modeled, from its manufacture to its end-of-life treatment, following the usual steps defined by the ISO 14040 standards (ISO, 2006).

Materials, goal and scope

As previously mentioned, we focus on carbon/epoxy composites. The deposit of materials to be recycled consists possibly in end-of-life aeronautic parts, but most likely to date, in composite offcuts. The composite part chosen for the LCA is assumed to be processed in Europe, with a Japanese carbon reinforcement. Its mass is supposed to be 1 kg. Thus, we aim at studying the interest of recycling such materials, that is to say more generally, the viability of the recycling process.

Life cycle inventory

The following analysis is based on Duflou *et al.*'s data (Duflou *et al.*, 2009), who assessed the manufacturing of composite semi-structural panels in automotive industry. All these data have been recalculated relative to the mass of the chosen product (*i.e.* 1 kg).

In our case study, the use phase is not taken into account. Indeed, the only input data concern transport operations. Like so, as rather classically, the present simulation led to show that this factor did not contribute much to the overall impacts (less than 5%).

Regarding the product's end-of-life, two scenarios have been modeled:

- the first one consists in burying the composite part, that is what is currently done;
- the second one consists in the recovery of the carbon reinforcement. The recycling process we focus on has been developed at laboratory scale (ICMCB, Bordeaux). It consists in
    (i) an aqueous solvolysis of the matrix by water under supercritical conditions (temperature around 400°C and pressure about 250 bar);
    (ii) and a hydrothermal oxidation of the effluent to clear matrix components from water, at the end of the solvolysis process.

  This technology is the only one that allows the fiber to be recovered. Therefore, it is a real (but partial) recycling, and not a simple material valorization (Morin, Loppinet-Serani, Cansell, & Aymonier, 2012). Lastly, the device uses energy, water and oxygen, and only emits water and carbon dioxide.

Lastly, O. Mantaux and A. Gillet from the MPI Department of the Mechanics Institute of Bordeaux, have developed a prototype for packaging these second-generation fibers in an attractive form for users (*i.e.* designers). Data matching the remanufacturing stage have not been taken into account yet in this very first LCA. However, this energy input is assumed to be very weak, compared to those involved in the first-generation reinforcement process. As a consequence, the life cycle only loops after the manufacturing of the first-generation carbon reinforcement, with no specific additional remanufacturing.

Method
The LCA is led with the SimaPro software and the ReCiPe Midpoint (H) method (Goedkoop *et al.*, 2012). As previously mentioned, in the recycling stage, the skipped material is the reinforcement. In other words, the production of a new raw material with non-renewable resources (*i.e.* first-generation carbon reinforcement), is avoided.

RESULTS AND DISCUSSION

Environmental validation
The LCA of a 1 kg composite part that takes into account the recycling of the reinforcement, clearly shows the interest of this end-of-life option. Actually, it almost offsets the whole environmental impacts of the composite manufacturing (cf. Figure 2). For example, emission of greenhouse gases may be divided by 10, compared to the landfill option, despite electricity consumption in the recycling process. The environmental gain is on average about 70%, according to the ReCiPe Midpoint (H) method.

Economic validation
In 2010, a market study led by Alcimed has shown that there will always be relevant uses for

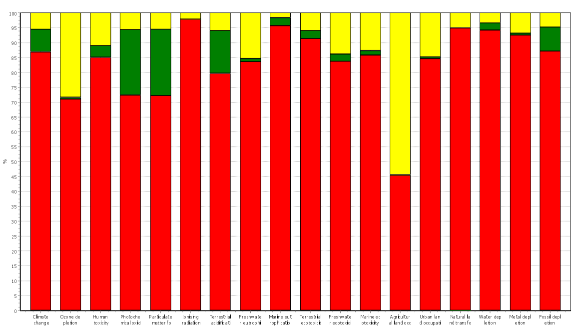

Figure 1. Environmental impacts due to the carbon reinforcement (*red*), the epoxy matrix (*green*) and the injection molding process (*yellow*), while processing a 1 kg carbon/epoxy composite part. The analysis is based on Duflou *et al.* (2009) data and on the ReCiPe Midpoint (H) method.

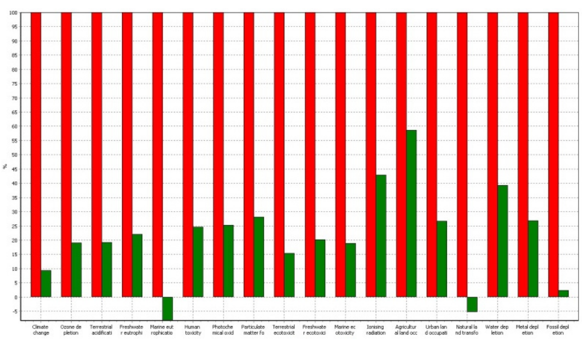

Figure 2. Life-cycle impact assessment of the landfill of a 1 kg carbon/epoxy composite part (*red*), compared with the reinforcement's recycling (*green*). The analysis is based on the ReCiPe Midpoint (H) method.

recycled reinforcements or for semi-products based on second-generation fiber, whatever their mechanical characteristics are, and as long as the price remains reasonable (Alcimed, 2010). The integration of recycled carbon fiber is only interesting if the mechanical performances/price ratio is higher than glass fiber's. Therefore, in light of excellent second-generation reinforcement's mechanical properties (Mantaux, Aymonier, & Antal, 2009), this ratio should be much higher than for new carbon fibers. Thus, the viability of the recycling will be provided if the second-generation semi-products price does not exceed 70-80% of the new ones.

CONCLUSIONS

In the present context, the use of carbon/epoxy composite is ever increasing. Now, it is well known that those composites can be recycled (Morin *et al.*, 2012), keeping good mechanical properties (Mantaux *et al.*, 2009). Taking right now into account that they will be soon subjected to regulation, it is essential to show that the composite recycling network will be viable, both economically and environmentally.

The recovery of the carbon reinforcement (which is the most environmentally impacting constituent in the composite manufacturing) by an aqueous solvolysis of the composite's matrix, leads to an average gain of about 70% for all eco-indicators compared to the landfill end-of-life option.

Lastly, the remanufacturing process developed by the Mechanics Institute of Bordeaux allows obtaining a semi-product easily usable. Consequently, from an economic point of view, the mechanical performances/price ratio of the second-generation carbon fiber should be higher than the virgin carbon fiber's, or the glass reinforcement's one.

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